

# Magnon pairing in quantum spin nematic

M. E. ZHITOMIRSKY<sup>1,2,3</sup> and H. TSUNETSUGU<sup>2</sup>

<sup>1</sup> *Service de Physique Statistique, Magnétisme et Supraconductivité, UMR-E9001 CEA-INAC/UJF  
17 rue des Martyrs, F-38054 Grenoble cedex 9, France*

<sup>2</sup> *Institute for Solid State Physics, University of Tokyo, Kashiwanoha, 5-1-5, Chiba 277-8581 Japan*

<sup>3</sup> *Max-Planck-Institut für Physik Komplexer Systeme, Nöthnitzer str. 38, D-01187 Dresden, Germany*

PACS 75.10.Jm – Quantized spin models, including quantum spin frustration

PACS 75.10.Kt – Quantum spin liquids, valence bond phases and related phenomena

PACS 75.10.Pq – Spin chain models

**Abstract.** – Competing ferro- and antiferromagnetic exchange interactions may lead to the formation of bound magnon pairs in the high-field phase of a frustrated quantum magnet. With decreasing field, magnon pairs undergo a Bose-condensation prior to the onset of a conventional one-magnon instability. We develop an analytical approach to study the zero-temperature properties of the magnon-pair condensate, which is a bosonic analog of the BCS superconductors. Representation of the condensate wave-function in terms of the coherent bosonic states reveals the spin-nematic symmetry of the ground-state and allows one to calculate various static properties. Sharp quasiparticle excitations are found in the nematic state with a small finite gap. We also predict the existence of a long-range ordered spin-nematic phase in the frustrated chain material  $\text{LiCuVO}_4$  at high fields.

**Introduction.** – Frustrated spin systems are interesting in general, as their zero- and low-temperature properties are governed by quantum fluctuations. These strong fluctuations may inhibit the formation of long-range magnetic order, stabilizing instead a disordered spin liquid. Following Anderson [1], much of the interest in the past decades has been focused on the investigation of various quantum spin-liquid states [2,3]. Another interesting possibility is the appearance of unconventional magnetic order characterized by partial breaking of the spin-rotational symmetry  $O(3)$ . A specific example is provided by spin-nematic states, which are analogous to the ordered phases of needle-like molecules in liquid crystals. Spin-nematic phases have been discussed phenomenologically in [4, 5], whereas identification of the relevant microscopic mechanism remains a challenging theoretical problem.

It was suggested long time ago [6] that a sizable biquadratic exchange  $(\mathbf{S}_i \cdot \mathbf{S}_j)^2$  in magnetic insulators with  $S \geq 1$  may stabilize a quadrupolar phase with vanishing sublattice magnetization  $\langle \mathbf{S} \rangle = 0$ , but a nonzero second-rank tensor, *e.g.*,  $\langle S_x^2 \rangle \neq \langle S_y^2 \rangle = \langle S_z^2 \rangle$ . In most real compounds the biquadratic exchange is, however, rather small. Recently, the interest in the biquadratic mechanism for the spin-nematic order has been revived in connection with the experiments on cold atomic gases [7, 8] and on the disor-

dered magnetic material  $\text{NiGa}_2\text{S}_4$  [9–11].

In this Letter we explore an alternative mechanism for the spin-nematic ordering based on competition between ferro- and antiferromagnetic interactions in magnetic insulators with an arbitrary value of the local spin including  $S = 1/2$ . Specifically, the mechanism operates in strong magnetic field and is based on the formation of bound magnon pairs in the fully polarized state [12–15]. This scenario has been studied numerically in a number of works on the so-called ferromagnetic  $J_1$ – $J_2$  chain model [16–20] and its generalization to two dimensions (2D) [21, 22]. Clear numerical evidence was found for the critical quadrupolar correlations below the saturation field for the 1D case. Note, however, that there is no true long-range order, nematic and otherwise, in one-dimensional quantum magnets at zero temperature. Therefore, a number of important questions on stability of the ordered nematic state and its excitation spectra are not answered by studying the purely 1D model.

The purpose of this Letter is to develop a simple analytical framework to treat the ground-state properties and low-energy magnetic excitations in the phase with a long-range spin-nematic order. Our description of the condensate of bound magnon pairs resembles in many aspects the BCS theory for the condensate of bound electron pairs

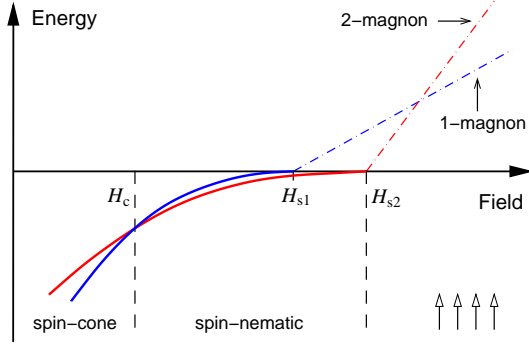


Fig. 1: (Color online) Energy-field diagram for a frustrated quantum magnet close to the saturation field. Dot-dashed lines show lowest one- and two-magnon states. Solid lines represent the ground state energy for the one-magnon (spin-cone) and the two-magnon (spin-nematic) condensate.

in superconductors. In addition, we predict that a spin-nematic phase must exist at high fields in the frustrated chain material  $\text{LiCuVO}_4$  [23–28].

The phenomenon of magnon pair condensation has a close relationship to the old problem of particle versus pair-superfluidity in an attractive Bose gas [29]. A common outcome is the density collapse prior to the pair-condensation transition [30]. Unrestricted growth of the local magnon density in spin-1/2 antiferromagnets is cured by their hard-core repulsion. In addition, reduced dimensionality of a spin subsystem found in many real materials helps to stabilize bound pairs and creates favorable conditions for their condensation. Experiments on  $\text{LiCuVO}_4$  and other related compounds in high magnetic fields may, therefore, lead to the first observation of such an exotic off-diagonal long-range order in solid-state systems.

In order to demonstrate the occurrence of magnon pair condensation at high magnetic fields we start with a general quantum Heisenberg antiferromagnet on an  $N$ -site lattice

$$\hat{H} = \frac{1}{2} \sum_{i,\mathbf{r}} J(\mathbf{r}) \mathbf{S}_i \cdot \mathbf{S}_j - H \sum_i S_i^z, \quad (1)$$

where  $\mathbf{r} = \mathbf{r}_j - \mathbf{r}_i$ . In the following  $S = 1/2$  is set for definiteness. In strong magnetic fields the Zeeman energy dominates over the exchange interactions and stabilizes the fully polarized state  $|0\rangle = |\uparrow\uparrow\uparrow\dots\rangle$ . This state is the vacuum for single spin-flips or magnons

$$|1_{\mathbf{q}}\rangle = \frac{1}{\sqrt{N}} \sum_i e^{-i\mathbf{q}\mathbf{r}_i} S_i^- |0\rangle \quad (2)$$

with the excitation energy

$$\varepsilon_{\mathbf{q}} = H + \frac{1}{2} \sum_{\mathbf{r}} J(\mathbf{r}) [e^{i\mathbf{q}\mathbf{r}} - 1] = H + \frac{1}{2} (J_{\mathbf{q}} - J_0), \quad (3)$$

where  $J_{\mathbf{q}} = \sum_{\mathbf{r}} J(\mathbf{r}) e^{i\mathbf{q}\mathbf{r}}$ . In ordinary antiferromagnets spin-flips repel each other. Then, once the band gap in  $\varepsilon_{\mathbf{q}}$

vanishes at a certain  $\mathbf{Q}$ ,  $J_{\mathbf{Q}} \equiv \min\{J_{\mathbf{q}}\}$ , an antiferromagnet undergoes a second-order transition into a canted spin structure at the saturation field

$$H_{s1} = \frac{1}{2} (J_0 - J_{\mathbf{Q}}), \quad (4)$$

as illustrated in Fig. 1. The antiferromagnetic state below  $H_{s1}$  can be regarded as a Bose-condensate of single magnons [31, 32].

**Two-magnon bound states.** – The conventional scenario for an antiferromagnetic transition in a strong magnetic field may change if some of the exchange bonds are ferromagnetic. In this case two spin-flips occupying the same bond with  $J(\mathbf{r}) < 0$  lower their interaction energy and may form a bound pair [12, 13]. To treat the bound state problem we follow the standard approach [33–35] and define a general two-magnon state

$$|2\rangle = \frac{1}{2} \sum_{i,j} f_{ij} S_i^- S_j^- |0\rangle, \quad (5)$$

with  $f_{ij} = f_{ji}$  being the magnon pair wave-function. Separating the center of mass motion  $f_{ij} = e^{i\mathbf{k}(\mathbf{r}_i + \mathbf{r}_j)/2} f_{\mathbf{k}}(\mathbf{r})$  and calculating the matrix elements of  $\hat{H}$  for states (5) we obtain the Bethe-Salpeter equation

$$(\varepsilon_2 - \varepsilon_{\mathbf{k}/2+\mathbf{q}} - \varepsilon_{\mathbf{k}/2-\mathbf{q}}) f_{\mathbf{k}}(\mathbf{q}) = \frac{1}{2N} \sum_{\mathbf{p}} (J_{\mathbf{p}+\mathbf{q}} + J_{\mathbf{p}-\mathbf{q}} - J_{\mathbf{k}/2+\mathbf{q}} - J_{\mathbf{k}/2-\mathbf{q}}) f_{\mathbf{k}}(\mathbf{p}), \quad (6)$$

where  $f_{\mathbf{k}}(\mathbf{q})$  is the Fourier transform of  $f_{\mathbf{k}}(\mathbf{r})$  and  $\varepsilon_2$  measures the magnon pair energy relative to the energy of the ferromagnetically polarized state. The above equation extends the previous theories [12, 13, 15] to an arbitrary geometry of exchange interactions and with a trivial replacement of  $\varepsilon_{\mathbf{q}}$  it also remains valid for an arbitrary value of spin  $S$ .

While the subsequent theoretical consideration is entirely general, we introduce now for illustration a specific spin model shown in Fig. 2, which is related to the quasi-1D helical antiferromagnet  $\text{LiCuVO}_4$ . This material consists of planar arrays of spin-1/2 copper chains with a ferromagnetic nearest-neighbor exchange  $J_1 < 0$  and an antiferromagnetic second-neighbor coupling  $J_2 > 0$ . Chains are linked by diagonal bonds, whereas interplanar interactions are an order of magnitude smaller. Neutron scattering measurements provide the following estimate for the exchange parameters in  $\text{LiCuVO}_4$ :  $J_1 = -1.6$  meV,  $J_2 = 3.8$  meV, and  $J_3 = -0.4$  meV [24]. The importance of quantum effects in this material is revealed by a small value of ordered moments  $\sim 0.3\mu_B$  in zero magnetic field [23]. The purely 1D  $J_1$ - $J_2$  model has been studied intensively in the past [12, 16–20] though no results exist for a realistic planar model.

To solve the integral equation (6) we expand  $f_{\mathbf{k}}(\mathbf{q})$  into lattice harmonics and obtain a finite algebraic system.

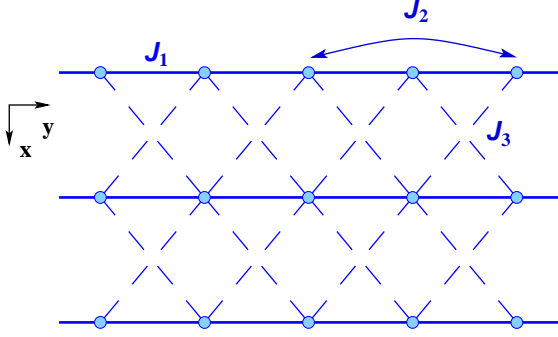


Fig. 2: (Color online) Two dimensional array of copper ions in LiCuVO<sub>4</sub> with principal exchange couplings.

Bound states exist for  $0.8\pi \leq k_y \leq \pi$  with the minimum of  $\varepsilon_2(\mathbf{k})$  at  $\mathbf{K} = (\pi, \pi)$ . The binding energy defined by  $\varepsilon_2(\mathbf{K}) = 2\varepsilon_{\mathbf{Q}} - E_B$  is found numerically to be  $E_B \approx 0.030J_2$ . In the absence of bound states the two-magnon continuum has a gap that is twice larger than the lowest one-magnon energy  $\varepsilon_{\mathbf{Q}} = H - H_{s1}$ . The two gaps vanish, therefore, in the same magnetic field. When bound states are present, condensation of magnon pairs starts in a higher magnetic field:

$$H_{s2} = H_{s1} + \frac{1}{2} E_B, \quad (7)$$

see Fig. 1. The condensation field for LiCuVO<sub>4</sub> is calculated to be  $H_{s2} = 47.1$  T ( $g = 2$ ), whereas the single magnon branch softens at  $H_{s1} = 46.5$  T. The relation  $H_{s2} > H_{s1}$  holds up to  $J_3 \approx -0.6$  meV. Hence, the conclusion about the magnon pair condensation in LiCuVO<sub>4</sub> is rather robust and should not be affected by a possible uncertainty in the experimental coupling constants [24].

We finish our analysis of the linear problem by presenting the wave-function of the lowest energy bound pairs in the momentum representation:

$$f_{\mathbf{K}}(\mathbf{q}) = \frac{\lambda J_2 \cos q_y}{2J_2(1 - \cos 2q_y) + 4J_3 \sin q_x \sin q_y + \delta\varepsilon}, \quad (8)$$

where a numerical constant  $\delta\varepsilon = 0.130J_2$  is related to the binding energy and  $\lambda$  is the normalization factor. In real space the function  $e^{i\mathbf{K}\mathbf{r}/2} f_{\mathbf{K}}(\mathbf{r})$  has the odd parity under the reflection  $y \rightarrow -y$  and vanishes, therefore, at  $\mathbf{r} = 0$ .

**Coherent condensate of magnon pairs.** – Below  $H_{s2}$  the bound magnon pairs acquire negative energy and start to condense. It is convenient at this point to transform from spin-1/2 operators to the Holstein-Primakoff bosons:

$$S_i^z = \frac{1}{2} - a_i^\dagger a_i, \quad S_i^- = a_i^\dagger \sqrt{1 - a_i^\dagger a_i}, \quad (9)$$

expanding subsequently square-roots to the first order in  $a_i^\dagger a_i$ .

The many-body state with a macroscopic number of the lowest-energy pairs below  $H_{s2}$  can be expressed as the coherent boson state of the pair creation operator:

$$|\Delta\rangle = e^{-N|\Delta|^2/2} \exp\left[\frac{1}{2} \Delta \sum_{i,j} f_{ij} a_i^\dagger a_j^\dagger\right] |0\rangle. \quad (10)$$

Here  $f_{ij} = e^{i\mathbf{K}(\mathbf{r}_i + \mathbf{r}_j)/2} f_{\mathbf{K}}(\mathbf{r})$  is the wave-function of the lowest energy pairs and  $\Delta$  is the complex amplitude of the condensate. The state (10) is a bosonic equivalent of the BCS pairing wave-function for fermions and must be regarded as a variational ansatz, which can be further improved by taking into account pair-pair correlations.

We use the ground-state wave-function (10) to compute simple boson averages:

$$\begin{aligned} \langle a_{\mathbf{q}} \rangle &= 0, \quad \langle a_{\mathbf{K}/2+\mathbf{q}} a_{\mathbf{K}/2-\mathbf{q}} \rangle = \frac{\Delta f_{\mathbf{K}}(\mathbf{q})}{1 - |\Delta|^2 f_{\mathbf{K}}^2(\mathbf{q})}, \\ n_{\mathbf{K}/2+\mathbf{q}} &= \langle a_{\mathbf{K}/2+\mathbf{q}}^\dagger a_{\mathbf{K}/2+\mathbf{q}} \rangle = \frac{|\Delta|^2 f_{\mathbf{K}}^2(\mathbf{q})}{1 - |\Delta|^2 f_{\mathbf{K}}^2(\mathbf{q})} \end{aligned} \quad (11)$$

as well as a more complicated four-boson correlator:

$$\begin{aligned} \langle a_{\mathbf{p}/2+\mathbf{q}}^\dagger a_{\mathbf{p}/2-\mathbf{q}}^\dagger a_{\mathbf{p}/2+\mathbf{q}'} a_{\mathbf{p}/2-\mathbf{q}'} \rangle &= |\Delta|^2 \delta_{\mathbf{p},\mathbf{K}} \\ &\times (1 + n_{\mathbf{K}/2+\mathbf{q}} + n_{\mathbf{K}/2+\mathbf{q}'} ) \frac{f_{\mathbf{K}}(\mathbf{q}) f_{\mathbf{K}}(\mathbf{q}')}{1 - |\Delta|^4 f_{\mathbf{K}}^2(\mathbf{q}) f_{\mathbf{K}}^2(\mathbf{q}')} \\ &+ |\Delta|^4 (\delta_{\mathbf{q},\mathbf{q}'} + \delta_{\mathbf{q},-\mathbf{q}'} ) (1 + n_{\mathbf{p}/2+\mathbf{q}} + n_{\mathbf{p}/2-\mathbf{q}} ) \\ &\times \frac{f_{\mathbf{K}}^2(\frac{\mathbf{p}-\mathbf{K}}{2} + \mathbf{q}) f_{\mathbf{K}}^2(\frac{\mathbf{p}-\mathbf{K}}{2} - \mathbf{q})}{1 - |\Delta|^4 f_{\mathbf{K}}^2(\frac{\mathbf{p}-\mathbf{K}}{2} + \mathbf{q}) f_{\mathbf{K}}^2(\frac{\mathbf{p}-\mathbf{K}}{2} - \mathbf{q})}. \end{aligned} \quad (12)$$

From these one can derive various spin correlators. In particular, the absence of the single-magnon condensate  $\langle a_{\mathbf{q}} \rangle = 0$  translates into

$$\langle S_i^{x,y} \rangle = 0, \quad (13)$$

which signifies a lack of a usual antiferromagnetic order parameter. The transverse and longitudinal spin correlations are given in the leading order by

$$\langle S_i^- S_j^+ \rangle \approx |\Delta|^2 \sum_l f_{il}^* f_{lj}, \quad \langle \delta S_i^z \delta S_j^z \rangle \approx |\Delta|^2 |f_{ij}|^2 \quad (14)$$

with  $\delta S_i^z = S_i^z - \langle S_i^z \rangle$ . In accordance with the behavior of the bound-state wave-function  $f_{\mathbf{K}}(\mathbf{r})$  the two correlators decay exponentially as  $|\mathbf{r}_i - \mathbf{r}_j| \rightarrow \infty$ . The transverse magnetic structure factor  $S^\perp(\mathbf{q})$  has no Bragg peaks and exhibits diffuse liquid-like spin correlations with a characteristic shape in momentum space determined by the bound-state wave-function:

$$\langle \mathbf{S}_{\mathbf{q}}^\perp \cdot \mathbf{S}_{-\mathbf{q}}^\perp \rangle \approx |\Delta|^2 \left[ f_{\mathbf{K}}^2(\frac{\mathbf{K}}{2} + \mathbf{q}) + f_{\mathbf{K}}^2(\frac{\mathbf{K}}{2} - \mathbf{q}) \right]. \quad (15)$$

The longitudinal response  $S^{zz}(\mathbf{q})$  being formally of the same order in  $|\Delta|$  appears to be much weaker than  $S^\perp(\mathbf{q})$  in the vicinity of the saturation field  $H_{s2}$ .

The general definition of the spin nematic order parameter in the  $O(2)$ -symmetric case is

$$Q_{ij}^{\alpha\beta} = \frac{1}{2} \langle S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha \rangle - \frac{1}{2} \delta_{\alpha\beta} \langle \mathbf{S}_i^\perp \cdot \mathbf{S}_j^\perp \rangle \quad (16)$$

where  $i, j$  belong to a nearest-neighbor bond and  $\alpha, \beta = x, y$ . The quadrupolar tensor  $Q_{ij}^{\alpha\beta}$  acquires a nonzero expectation value in the presence of the pair condensate:

$$Q_{ij}^{xx} + iQ_{ij}^{xy} = \frac{1}{2} \langle S_i^+ S_j^+ \rangle \approx \frac{\Delta}{2} f_{ij}. \quad (17)$$

The phase of the condensate amplitude  $\Delta$  determines the orientation of the spin-nematic director in the  $x$ - $y$  plane. The director forms a periodic structure in the real space determined by the momentum  $\mathbf{K}$ .

Using explicit expressions for the spin correlators, one can calculate the ground-state energy. Below, we shall use for this purpose a complementary approach, which is analogous to the Bogoliubov method in the theory of superconductivity and yields the spectrum of quasiparticle excitations simultaneously with the static properties.

**Mean-field approach.** – The bosonic equivalent of the spin Hamiltonian is obtained upon substitution of (9) into (1). We restrict ourselves to quadratic and quartic terms in  $a_i$ 's and define two types of mean-field averages for each exchange bond:

$$\Delta_{ij} = \langle a_i a_j \rangle, \quad n_{\mathbf{r}} = \langle a_i^\dagger a_i \rangle, \quad (18)$$

and the magnon density  $n = \langle a_i^\dagger a_i \rangle$ . The anomalous correlator is further factorized as  $\Delta_{ij} = e^{i\mathbf{K}(\mathbf{r}_i + \mathbf{r}_j)/2} \Delta_{\mathbf{r}}$ . Both  $\Delta_{\mathbf{r}}$  and  $n_{\mathbf{r}}$  are even real functions of  $\mathbf{r}$  with a proper choice of gauge. Performing the mean-field decoupling in the interaction term we obtain a quadratic form, which is then diagonalized with the canonical transformation. This yields the energy of one-magnon excitations

$$\begin{aligned} \epsilon_{\mathbf{K}/2+\mathbf{q}} &= \epsilon_{\mathbf{q}} - \sum_{\mathbf{r}} J(\mathbf{r}) \left( \frac{1}{2} - n - n_{\mathbf{r}} \right) \sin \frac{1}{2} \mathbf{K} \mathbf{r} \sin \mathbf{q} \mathbf{r}, \\ \epsilon_{\mathbf{q}} &= \sqrt{A_{\mathbf{q}}^2 - B_{\mathbf{q}}^2}, \quad B_{\mathbf{q}} = \sum_{\mathbf{r}} J(\mathbf{r}) \Delta_{\mathbf{r}} \cos \mathbf{q} \mathbf{r}, \quad (19) \\ A_{\mathbf{q}} &= H - \sum_{\mathbf{r}} J(\mathbf{r}) \left( \frac{1}{2} - n - n_{\mathbf{r}} \right) (1 - \cos \frac{1}{2} \mathbf{K} \mathbf{r} \cos \mathbf{q} \mathbf{r}). \end{aligned}$$

In accordance with the exponential decay of spin correlations (14), the excitation spectrum acquires a gap in the presence of the magnon pair condensate. The above expressions are used to calculate bosonic averages and to obtain a closed form of the self-consistent equations:

$$\Delta_{\mathbf{r}} = \sum_{\mathbf{q}} \frac{B_{\mathbf{q}}}{2\epsilon_{\mathbf{q}}} \cos \mathbf{q} \mathbf{r}, \quad n_{\mathbf{r}} = \sum_{\mathbf{q}} \frac{A_{\mathbf{q}}}{2\epsilon_{\mathbf{q}}} \cos(\frac{1}{2} \mathbf{K} + \mathbf{q}) \mathbf{r}. \quad (20)$$

In the limit  $H \rightarrow H_{s2}$  one finds  $\Delta_{\mathbf{r}} \gg n_{\mathbf{r}} \sim \Delta_{\mathbf{r}}^2$ , while the linearized equation for  $\Delta_{\mathbf{r}}$  transforms directly into the bound-state equation (6).

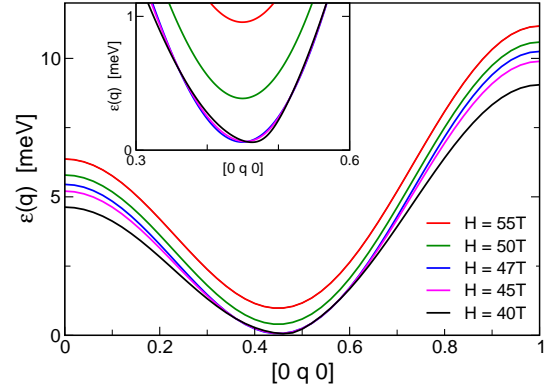


Fig. 3: (Color online) Dispersion of a single-magnon branch in LiCuVO<sub>4</sub> in the fully polarized state ( $H_{s2} \approx 47$  T) and in the state with the magnon pair condensate. Field values for different curves from top to bottom are listed on the plot. The inset shows the vicinity of the magnon gap.

We have solved self-consistently the set of equations (19) and (20) and calculated the ground-state energy for the spin model of Fig. 2 assuming the same symmetry of bond variables  $\Delta_{\mathbf{r}}$  and  $n_{\mathbf{r}}$  as in the magnon pair coherent state (10). With decreasing external field, the pairs overlap more appreciably and at a certain point give way to a conventional one-particle condensation. Comparing the ground-state energy of the pair condensate with the energy of a simple spin-cone structure we find the first-order transition at  $H_c \approx 44.5$  T as illustrated schematically in Fig. 1. The spin-nematic state has the lowest energy in a finite range of fields  $H_c < H < H_{s2}$ , which extends well below the condensation field  $H_{s1}$  for single magnons.

The ground-state energy calculation allows to determine the slope of the magnetization curve  $M(H)$  at  $H_c < H < H_{s2}$ . In ordinary quasi-1D antiferromagnets,  $M(H)$  deviates from a straight line as  $H \rightarrow H_s$  resembling the square-root singularity of a single quantum spin chain. Our mean-field calculations for the high-field nematic phase in LiCuVO<sub>4</sub> yield instead the slope  $dM/dH \approx 0.38 M_{\text{sat}}/J_2$ , which amounts to only 54% of the slope of the classical magnetization curve. The quantum corrections beyond the mean-field approximation should somewhat modify this value. However, we expect them to be small for the same reason as the suppression of critical fluctuations in the BCS superconductors. Indeed, the size of the bound magnon pairs is rather large extending to  $\xi \sim 10$  interatomic spacing in the direction of chains. Already for small magnon densities each bound pair is surrounded by many neighboring magnon pairs, which enforces the mean-field behavior. Therefore, a distinct signature of the high-field nematic phase in LiCuVO<sub>4</sub> will be a sharp change in the slope of the magnetization curve.

The dispersion of one-magnon excitations found together with the ground-state energy in the self-consistent calculation is presented in Fig. 3. In the fully polarized state at  $H > H_{s2} \approx 47$  T, variation of the applied field

results in an overall shift of the magnon energy according to Eq. (3). At  $H = H_{s2}$  magnons have a small gap  $\Delta_g = H_{s2} - H_{s1} \approx 0.06$  meV. The field dependence of  $\varepsilon_{\mathbf{k}}$  changes drastically in the presence of the magnon pair condensate. Decreasing the field modifies the shape of  $\varepsilon_{\mathbf{k}}$  but the excitation gap  $\Delta_g$  remains practically unchanged, see the inset in Fig. 3. The lowest field  $H = 40$  T used in Fig. 3 is below the transition field  $H_c$  into the spin-cone magnetic structure. The corresponding dispersion curve  $\varepsilon_{\mathbf{k}}$  illustrates that the spin-nematic state remains locally stable even below  $H_c$ . This is in contrast with the previous scenario suggested for an attractive Bose gas, for which the pair condensate was assumed to become unstable due to a softening of the single-particle branch [29, 30].

The motion of the spin-nematic order parameter provides an additional gapless branch of collective excitations, which should yield a nonzero dynamical signal in the longitudinal channel. Transverse spin-spin correlations in the nematic phase are dominated by unpaired magnons (19).

Finally, let us also comment on a low-field state at  $H < H_c$ . The considered scenario of a direct transition between the spin-nematic state and the conventional canted antiferromagnetic phase applies most certainly to the 2D model [21], which exhibits a magnetic ordering at  $\mathbf{Q} = (\pi, 0)$  in zero field. In the case of weakly coupled chains, the low-field phase might be more complicated than the simple conical spin-structure used above as an example. The NMR measurements [26, 28] indicate that the intermediate-field phase, which is observed in  $\text{LiCuVO}_4$  above 8 T [25], has predominant longitudinal SDW-type correlations between local spins. This experimental finding agrees, in principle, with the numerical results for a single spin chain [17–19]. Understanding the fate of such a 1D phase in the presence of interchain couplings as well as its relation to the transverse nematic order remains an open theoretical problem.

To summarize, we have presented the analytical description for the Bose-condensate of bound magnon pairs in a frustrated quantum magnet in high magnetic fields. The theory applies to a number of real magnetic compounds with competing ferro- and antiferromagnetic interactions. After the first version of the present work has appeared, we learned about the experimental observation of a new phase in  $\text{LiCuVO}_4$  in the field range 41–44 T [36].

\*\*\*

We are grateful to M. Enderle, B. Fåk, M. Hagiwara, and L. Svistov for stimulating discussions. Part of this work has been performed within the Advanced Study Group Program on “Unconventional Magnetism in High Fields” at the Max-Planck Institute for the Physics of Complex Systems. H.T. acknowledges support by Grants-in-Aid for Scientific Research (No. 17071011 and No. 19052003) and by the Next-Generation Supercomputing Project, Nanoscience Program, MEXT of Japan.

## REFERENCES

- [1] ANDERSON P. W., *Mater. Res. Bull.*, **8** (1973) 153.
- [2] FRADKIN E., *Field Theories of Condensed Matter Systems* (Addison-Wesley, Reading) 1991.
- [3] MISGUICH G. and LHUILLIER C., in *Frustrated Spin Systems*, edited by DIEP H. T. (World Scientific, Singapore) 2005.
- [4] ANDREEV A. F. and GRISHCHUK I. A., *Sov. Phys. JETP*, **60** (1984) 267.
- [5] CHANDRA P. and COLEMAN P., *Phys. Rev. Lett.*, **66** (1991) 100.
- [6] BLUME M. and HSIEH Y. Y., *J. Appl. Phys.*, **40** (1969) 1249.
- [7] STENGER J. *et al.*, *Nature*, **396** (1998) 345.
- [8] DEMLER E. and ZHOU F., *Phys. Rev. Lett.*, **88** (2002) 163001.
- [9] NAKATSUJI S. *et al.*, *Science*, **309** (2005) 1697.
- [10] TSUNETSUGU H. and ARIKAWA M., *J. Phys. Soc. Jpn.*, **75** (2006) 083701.
- [11] LÄUCHLI A., MILA F., and PENC K., *Phys. Rev. Lett.*, **97** (2006) 087205.
- [12] CHUBUKOV A. V., *Phys. Rev. B*, **44** (1991) 4693.
- [13] KUZIAN R. O. and DRECHSLER S.-L., *Phys. Rev. B*, **75** (2007) 024401.
- [14] DMITRIEV D. V. and KRIVNOV V. YA., *Phys. Rev. B*, **79** (2009) 054421.
- [15] UEDA H. T. and TOTSUKA K., *Phys. Rev. B*, **80** (2009) 014417.
- [16] HEIDRICH-MEISNER F., HONECKER A. and VEKUA T., *Phys. Rev. B*, **74** (2006) 020403(R).
- [17] VEKUA T. *et al.*, *Phys. Rev. B*, **76** (2007) 174420.
- [18] HIKIHARA T. *et al.*, *Phys. Rev. B*, **78** (2008) 144404.
- [19] SUDAN J., LUSCHER A. and LÄUCHLI A. M., *Phys. Rev. B*, **80** (2009) 140402(R).
- [20] HEIDRICH-MEISNER F., MCCULLOCH I. P., and KOLEZHUK A. K., *Phys. Rev. B*, **80** (2009) 144417.
- [21] SHANNON N., MOMOI T. and SINDZINGRE P., *Phys. Rev. Lett.*, **96** (2006) 027213.
- [22] SHINDOU R. and MOMOI T., *Phys. Rev. B*, **80** (2009) 064410.
- [23] GIBSON B. J. *et al.*, *Physica B*, **350** (2004) e253.
- [24] ENDERLE M. *et al.*, *Europhys. Lett.*, **70** (2005) 237.
- [25] BANKS M. G. *et al.*, *J. Phys.: Cond. Mat.*, **19** (2007) 145227.
- [26] BUTTGEN N. *et al.*, *Phys. Rev. B*, **76** (2007) 014440.
- [27] SCHRETTLE F. *et al.*, *Phys. Rev. B*, **77** (2008) 144101.
- [28] BUTTGEN N. *et al.*, *Phys. Rev. B*, **81** (2010) 052403.
- [29] VALATIN J. G. and BUTLER D., *Nuovo Cimento*, **10** (1958) 37.
- [30] NOZIÈRES P. and SAINT JAMES D., *J. Phys. (Paris)*, **43** (1982) 1133.
- [31] MATSUBARA T. and MATSUDA H., *Prog. Theor. Phys.*, **16** (1956) 569.
- [32] BATYEV E. G. and BRAGINSKII L. S., *Sov. Phys. JETP*, **60** (1984) 781.
- [33] WORTIS M., *Phys. Rev.*, **132** (1963) 85; **138** (1965) A1126.
- [34] HANUS J., *Phys. Rev. Lett.*, **11** (1963) 336.
- [35] MATTIS D. C., *The Theory of Magnetism I* (Springer, Berlin) 1988.
- [36] SVISTOV L. E. *et al.*, [arXiv:1005.5668](https://arxiv.org/abs/1005.5668).